

Am

(CONVENTION. By one or more)

CC



## CONVENTION APPLICATION FOR A PATENT

(1) Here insert in full Name of Applicant or Applicants, followed by Address (es).

1 HOECHST AKTIENGESELLSCHAFT,  
We of 45 Bruningstrasse, D-6230 Frankfurt/Main 80,  
Federal Republic of Germany

(2) Here insert Title of Invention.

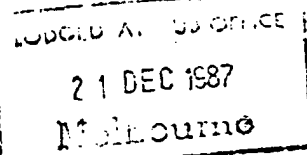
hereby apply for the grant of a Patent for an invention entitled:  
BICYCLIC IMIDES. A PROCESS FOR THEIR PREPARATION, AND  
THEIR USE IN PLANT PROTECTION

(3) Here insert number(s) of basic application(s).

which is described in the accompanying complete specification. This application is a Convention application and is based on the application numbered<sup>(3)</sup>  
P36 43 748.4

(4) Here insert Name of basic Country or Countries, and date of filing.

for a patent or similar protection made in<sup>(4)</sup> Federal Republic of Germany  
on 20th December 1986



My address for service is Messrs. Edwd. Waters & Sons, Patent Attorneys,  
Our 50 Queen Street, Melbourne, Victoria, Australia.

DATED this 18th day of December 1987

HOECHST AKTIENGESELLSCHAFT

*James Murray*  
James Murray

Registered Patent Attorney

BEST AVAILABLE COPY

COMMONWEALTH OF AUSTRALIAPatents Act 1952DECLARATION IN SUPPORT OF A CONVENTION APPLICATION UNDER PART XVI.  
FOR A PATENT.

In support of the Convention application made under Part XVI. of the Patents Act 1952 by HOECHST AKTIENGESELLSCHAFT of 45, Brüningstrasse, D-6230 Frankfurt/Main 80, Federal Republic of Germany for a patent for an invention entitled:

BICYCLIC IMIDES. A PROCESS FOR THEIR PREPARATION, AND THEIR USE IN PLANT PROTECTION

We, Bernhard Fock, of Brüsselweg, D-6230 Frankfurt/Main 80, Federal Republic of Germany, do solemnly and sincerely declare as follows:

1. We are authorized by HOECHST AKTIENGESELLSCHAFT the applicant for the patent to make this declaration on its behalf.

2. The basic application as defined by Section 141 of the Act was made in the Federal Republic of Germany under No. P 10 43 748.4 on December 1, 1957 by HOECHST AKTIENGESELLSCHAFT

3. a) Reinhold Liebl, of Am Alten Weidenweg, D-6230 Frankfurt/Main 80  
b) Michael Frey, of Marburger Strasse, D-6230 Frankfurt/Main 80  
c) Hilmar Mildenschnor, of Fasanenweg, D-6230 Frankfurt/Main 80  
d) Klaus Bauer, of Fasanenweg, D-6230 Frankfurt/Main 80  
e) Hermann Bieringer, of Fasanenweg, D-6230 Frankfurt/Main 80  
f) - of Fasanenweg, D-6230 Frankfurt/Main 80

is/are the actual inventor(s) of the invention and the facts upon which HOECHST AKTIENGESELLSCHAFT

is entitled to make the application are as follows:

The said HOECHST AKTIENGESELLSCHAFT

is the assignee of the said

Reinhold Liebl, Michael Frey, Hilmar Mildenschnor, Klaus Bauer, Hermann Bieringer

4. The basic application referred to in paragraph 2 of this Declaration was the first application made in a Convention country in respect of the invention the subject of the application.

DECLARED at Frankfurt/Main, Federal Republic of Germany

this 25th day of November 1957

To the Commissioner of Patents

HOECHST AKTIENGESELLSCHAFT

PAT 510

Prokurist Authorized Signatory

ppa. F. 2

I.V. Lapina

(12) PATENT ABSTRACT (11) Document No. AU-A-82875/87  
(19) AUSTRALIAN PATENT OFFICE

(54) Title  
BICYCLIC IMIDES

(51)4 International Patent Classification

C07D 471/04 C07F 009/65 C07D 487/04 C07D 498/04  
C07D 513/04 A61K 031/445 A61K 031/495

(21) Application No. : 82875/87 (22) Application Date : 21.12.87

(30) Priority Data

(31) Number (32) Date (33) Country  
3643748 20.12.86 DE FEDERAL REPUBLIC OF GERMANY

(43) Publication Date : 23.6.88

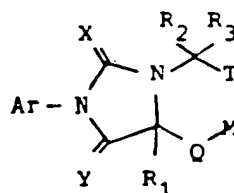
(71) Applicant  
HOECHST A.G.;

(72) Inventor  
RAINER LIEBL  
MICHAEL FREY  
HILMAR MILDENBERGER  
KLAUS BAUER  
HERMANN BIERINGER

(74) Attorney or Agent  
EDWD. WATERS & SONS

(57) Claim

1. A compound of the formula I, or a salt thereof,

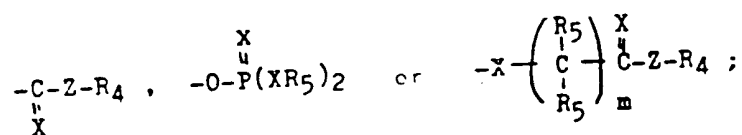


(I)

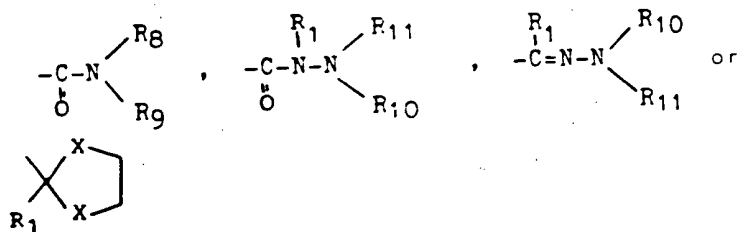
in which

Ar denotes phenyl, naphthyl, pyridyl, quinolinyl or isoquinolinyl, these radicals being monosubstituted to tetrasubstituted, preferably monosubstituted to trisubstituted, by identical or different radicals from the series comprising halogen, hydroxyl, (C<sub>1</sub>-C<sub>4</sub>)alkyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>6</sub>)cycloalkoxy, (C<sub>3</sub>-C<sub>6</sub>)alkenyloxy, (C<sub>3</sub>-C<sub>6</sub>)alkynyloxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxy-(C<sub>1</sub>-C<sub>4</sub>)-alkoxy, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxy, halo(C<sub>3</sub>-C<sub>6</sub>)alkenyloxy, halo(C<sub>3</sub>-C<sub>6</sub>)alkynyloxy, (C<sub>1</sub>-C<sub>4</sub>)alkylthio, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfinyl, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfonyl, NO<sub>2</sub>, -CN, -NHR<sub>1</sub>, cyano(C<sub>1</sub>-C<sub>4</sub>)alkyl, phenoxy, phenoxy(C<sub>1</sub>-C<sub>4</sub>)alkyl, phenyl(C<sub>1</sub>-C<sub>4</sub>)alkoxy, phenylthio, phenylsulfinyl or phenylsulfonyl, it being possible for the six lastmentioned radicals to

be monosubstituted, disubstituted or trisubstituted in the phenyl ring by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, -CN or NO<sub>2</sub>, or furthermore by a radical of the formula

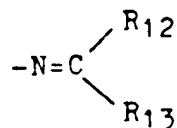


- R<sub>1</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or phenyl which may be monosubstituted or disubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, NO<sub>2</sub>, CN or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl,
- R<sub>2</sub> and R<sub>3</sub>, independently of one another, denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl which may be substituted by cyano, hydroxyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxy, phenyl or benzyl which may both be substituted, in each case preferably monosubstituted or disubstituted, in the phenyl ring by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, NO<sub>2</sub> or -CN; (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, hal. (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, carboxyl or a radical of the formula



- R<sub>4</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl which may be monosubstituted to hexasubstituted by halogen and/or monosubstituted or disubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxy(C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkylthio, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfinyl, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfonyl, (C<sub>1</sub>-C<sub>4</sub>)alkylamino, -CN, furyl, tetrahydrofuryl, benzofuryl, phenyl, phenoxy or

benzyloxy, it being possible for the six last-mentioned radicals to be monosubstituted to trisubstituted, in each case in the phenyl ring or the heteroaromatic ring, by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>6</sub>)cycloalkyl, (C<sub>3</sub>-C<sub>6</sub>)-alkenyl, cyclohexenyl, (C<sub>3</sub>-C<sub>6</sub>)alkynyl or phenyl, which may be monosubstituted to trisubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl; or a radical of the formula

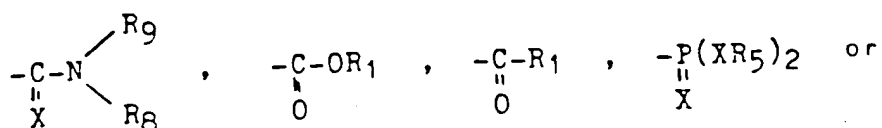


where the lastmentioned radical is excluded when  
Z = S,

R<sub>5</sub> denotes hydrogen or (C<sub>1</sub>-C<sub>4</sub>)alkyl,

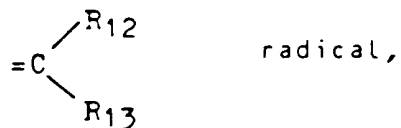
R<sub>6</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or, together with R<sub>4</sub> and the nitrogen atom connecting these radicals, a 5- to 7-membered heterocyclic ring which can contain as ring members one or two radicals from the series comprising -O-, -S- and -NR<sub>5</sub>- and which may be monosubstituted to trisubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkyl;

R<sub>7</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, phenyl or benzyl, where the phenyl ring may in each case be monosubstituted to trisubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, -NO<sub>2</sub>, CF<sub>3</sub>, -CN or a radical of the formula



$R_8$  and  $R_9$ , independently of one another, denote hydrogen or  $(C_1-C_4)$ alkyl, or the two radicals  $R_8$  and  $R_9$ , together with the nitrogen atom connecting them, denote a 5- to 7-membered heterocyclic ring which can contain as ring members one or two radicals from the series comprising  $-O-$ ,  $-S-$  and  $-NR_5-$  and which may be monosubstituted to trisubstituted by  $(C_1-C_4)$ alkyl, hydroxyl,  $(C_1-C_4)$ alkoxy, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in the phenyl ring by  $(C_1-C_4)$ alkyl,  $(C_1-C_4)$ alkoxy, halogen or  $(C_1-C_4)$ alkoxycarbonyl,

$R_{10}$  and  $R_{11}$ , independently of one another, denote hydrogen,  $(C_1-C_4)$ alkyl, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in each case in the phenyl ring, by halogen,  $(C_1-C_4)$ alkyl,  $(C_1-C_4)$ alkoxy,  $NO_2$ ,  $-CN$ ,  $CF_3$  or  $(C_1-C_4)$ alkoxycarbonyl, or  $R_{10}$  and  $R_{11}$  together denote the



$R_{12}$  and  $R_{13}$ , independently of one another, denote  $(C_1-C_4)$ alkyl,  $(C_1-C_4)$ alkoxycarbonyl, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in the phenyl ring by halogen,  $(C_1-C_4)$ alkyl,  $(C_1-C_4)$ alkoxy,  $(C_1-C_4)$ alkoxycarbonyl,  $CF_3$ ,  $-CN$  or  $NO_2$ ,

$M$  denotes  $\begin{array}{c} R_2 \\ \diagup \\ =C \\ \diagdown \\ R_3 \end{array}$ ,  $\begin{array}{c} O \\ | \\ -S- \\ | \\ O \end{array}$ ,  $\begin{array}{c} O \\ | \\ -S- \\ | \\ O \end{array}$ ,  $\begin{array}{c} O \\ | \\ -S- \\ | \\ O \end{array}$ ,  $-O-$  or  $NR_7$ ;

$Q$  denotes  $\begin{array}{c} R_2 \\ \diagup \\ =C \\ \diagdown \\ R_3 \end{array}$ ,  $-S-$  or  $-O-$ ,

- T denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{C} \\ \diagdown \\ \text{R}_3 \end{array}$ ,  $-\text{S}-$ ,  $\begin{array}{c} \text{O} \\ \parallel \\ -\text{S}- \end{array}$ ,  $\begin{array}{c} \text{O} \\ \parallel \\ -\text{S}- \\ \parallel \\ \text{O} \end{array}$  or  $-\text{O}-$ ,
- X denotes O or S,
- Y denotes O, S or NH,
- Z denotes O, S or NR<sub>6</sub>,
- m denotes 1, 2 or 3, and
- n denotes 1 or 2,
- with the proviso that m cannot be CH<sub>2</sub>, S, SO or SO<sub>2</sub> when Y denotes O, R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> denote H and Q and T denotes CH<sub>2</sub>.

5. The use of a compound of the formula I of claim 1 or 2, or a salt thereof, as a herbicide.

COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952-69

## COMPLETE SPECIFICATION

(ORIGINAL)

Class

Int. Class

Application Number:

Lodged:

Complete Specification Lodged:

Accepted:

Published:

Priority:

Related Art:

---

Name of Applicant: HOECHST AKTIENGESELLSCHAFTAddress of Applicant: 45 Bruningstrasse, D-6230 Frankfurt/Main 80,  
Federal Republic of GermanyActual Inventor: RAINER LIEBL, MICHAEL FREY, HILMAR MILDENBERGER, KLAUS  
BAUER and HERMANN BIERINGERAddress for Service: EDWD. WATERS & SONS,  
50 QUEEN STREET, MELBOURNE, AUSTRALIA, 3000.

Complete Specification for the invention entitled:

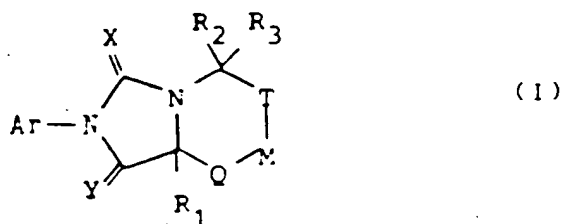
BICYCLIC IMIDES, A PROCESS FOR THEIR PREPARATION, AND  
THEIR USE IN PLANT PROTECTION

The following statement is a full description of this invention, including the best method of performing it known to US



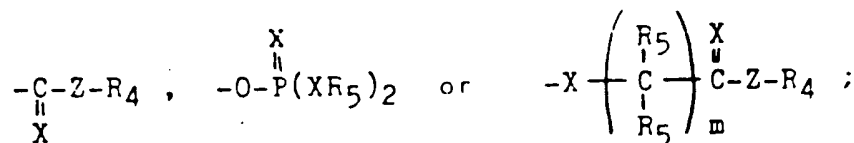
Bicyclic imides having a herbicidal activity are described in EP-A 70,389, EP-A 104,532 and US Patent 5 4,179,276.

The present invention therefore relates to compounds of  
10 the formula I, or their salts,



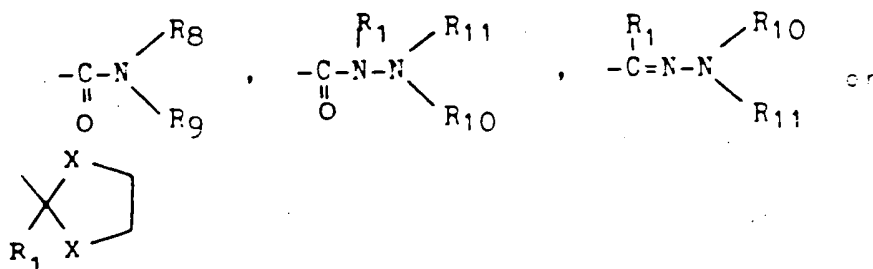
Ar denotes phenyl, naphthyl, pyridyl, quinolinyl or isoquinolinyl, these radicals being monosubstituted to tetrasubstituted, preferably monosubstituted to trisubstituted, by identical or different radicals from the series comprising halogen, hydroxyl, (C<sub>1</sub>-C<sub>4</sub>)alkyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>6</sub>)-cycloalkoxy, (C<sub>3</sub>-C<sub>6</sub>)alkenyloxy, (C<sub>3</sub>-C<sub>6</sub>)-alkynyloxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxy-(C<sub>1</sub>-C<sub>4</sub>)-alkoxy, halo-(C<sub>1</sub>-C<sub>4</sub>)alkoxy, halo(C<sub>3</sub>-C<sub>6</sub>)alkenyloxy, halo(C<sub>3</sub>-C<sub>6</sub>)-alkynyloxy, (C<sub>1</sub>-C<sub>4</sub>)alkylthio, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfinyl, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfonyl, NO<sub>2</sub>, -CN, -NHR<sub>1</sub>, cyano-(C<sub>1</sub>-C<sub>4</sub>)alkyl, phenoxy, phenoxy(C<sub>1</sub>-C<sub>4</sub>)alkyl, phenyl(C<sub>1</sub>-C<sub>4</sub>)alkoxy, phenylthio, phenylsulfinyl or phenylsulfonyl, it being possible for the six lastmentioned radicals to be monosubstituted, disubstituted or trisubstituted in the phenyl ring by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy,

(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, -CN or NO<sub>2</sub>. or furthermore by a radical of the formula



5 R<sub>1</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or phenyl which may be monosubstituted or disubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, NO<sub>2</sub>, CN or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl,

10 R<sub>2</sub> and R<sub>3</sub>, independently of one another, denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl which may be substituted by cyano, hydroxyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxy, phenyl or benzyl which may both be substituted, in each case preferably monosubstituted or disubstituted, in the phenyl ring by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, NO<sub>2</sub> or -CN;  
15 (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)-alkoxycarbonyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)-alkoxy(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)-alkoxycarbonyl(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, carbonyl or a radical of the formula



20 R<sub>4</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl which may be monosubstituted to hexasubstituted by halogen and/or monosubstituted or disubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxy(C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkylthio, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfinyl, 25 (C<sub>1</sub>-C<sub>4</sub>)alkylsulfonyl, (C<sub>1</sub>-C<sub>4</sub>)alkylamino, -CN, furyl, tetrahydrofuryl, benzofuryl, phenyl, phenoxy

or benzyloxy, it being possible for the six last-mentioned radicals to be monosubstituted to trisubstituted, in each case in the phenyl ring or the heteroaromatic ring, by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>6</sub>)cycloalkyl, (C<sub>3</sub>-C<sub>6</sub>)alkenyl, cyclohexenyl, (C<sub>3</sub>-C<sub>6</sub>)alkynyl or phenyl, which may be monosubstituted to trisubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl; or a radical of the formula

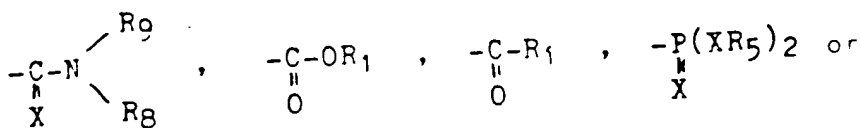


where the lastmentioned radical is excluded when Z = S,

R<sub>5</sub> denotes hydrogen or (C<sub>1</sub>-C<sub>4</sub>)alkyl,

R<sub>6</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or, together with R<sub>4</sub> and the nitrogen atom connecting these radicals, a 5- to 7-membered heterocyclic ring which can contain as ring members one or two radicals from the series comprising -O-, -S- and -NR<sub>5</sub>- and which may be monosubstituted to trisubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkyl;

R<sub>7</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, phenyl or benzyl, where the phenyl ring may in each case be monosubstituted to trisubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, -NO<sub>2</sub>, CF<sub>3</sub>, -CN or a radical of the formula

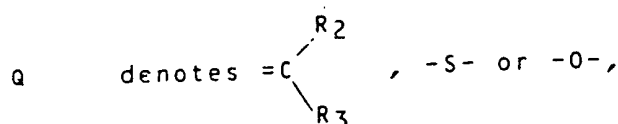
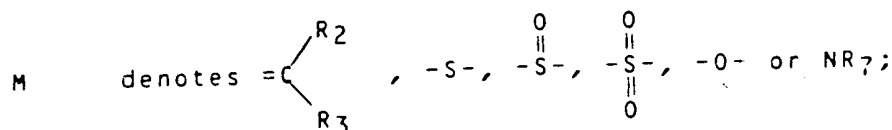


R<sub>8</sub> and R<sub>9</sub>, independently of one another, denote hydrogen or (C<sub>1</sub>-C<sub>4</sub>)alkyl, or the two radicals R<sub>8</sub> and R<sub>9</sub>, together with the nitrogen atom connecting them, denote a 5- to 7-membered heterocyclic ring which can contain as ring members one or two radicals from the series comprising -O-, -S- and -NR<sub>5</sub>- and which may be monosubstituted to trisubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkyl, hydroxyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in the phenyl ring by (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, halogen or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl,

R<sub>10</sub> and R<sub>11</sub>, independently of one another, denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in each case in the phenyl ring, by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, NO<sub>2</sub>, -CN, CF<sub>3</sub> or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, or R<sub>10</sub> and R<sub>11</sub> together denote the



R<sub>12</sub> and R<sub>13</sub>, independently of one another, denote (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in the phenyl ring by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, CF<sub>3</sub>, -CN or NO<sub>2</sub>,



T denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{C} \\ \diagdown \\ \text{R}_3 \end{array}$ ,  $-\text{S}-$ ,  $\begin{array}{c} \text{O} \\ \parallel \\ -\text{S}- \end{array}$ ,  $\begin{array}{c} \text{O} \\ \parallel \\ -\text{S}- \\ \parallel \\ \text{O} \end{array}$  or  $-\text{O}-$ ,

X denotes O or S,

Y denotes O, S or NH,

Z denotes O, S or  $\text{NR}_6$ ,

5 m denotes 1, 2 or 3, and

n denotes 1 or 2,

with the proviso that M cannot be  $\text{CH}_2$ , S, SO or  $\text{SO}_2$  when Y denotes O,  $\text{R}_1$ ,  $\text{R}_2$  and  $\text{R}_3$  denote H and Q and T denotes  $\text{CH}_2$ .

- 10 The compounds of the formula I can exist as pure stereoisomers or as mixtures thereof. All these isomeric forms are covered by the invention.

Salt formation in the case of the compounds of the formula I can take place when  $\text{R}_2$  or  $\text{R}_3$  denotes carboxyl or when  $\text{ZR}_4$  denotes OH or SH. Possible salts are generally those which can be employed in agriculture. These include, for example, the alkali metal salts and alkaline-earth metal salts, in particular Na, K, Mg and Ca salts, or the salts with ammonium, which may be monosubstituted to tetrasubstituted by organic radicals, in particular alkyl or hydroxyalkyl radicals.

In the definition of formula I, haloalkyl, haloalkoxy, haloalkenyloxy or haloalkynyloxy contains one or more halogen atoms, mainly one to six F, Cl or Br atoms.

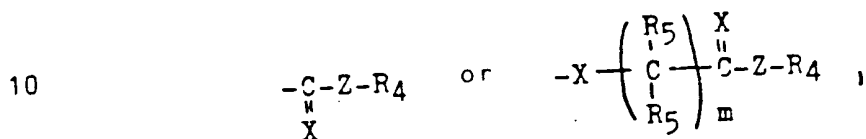
25 These include, for example, the  $-\text{CF}_3$ ,  $-\text{C}_2\text{F}_5$ ,  $-\text{CH}_2\text{CF}_3$ ,  $-\text{CF}_2\text{Cl}$ ,  $-\text{CF}_2\text{CHF}_2$ ,  $-\text{CF}_2\text{CHFCl}$ ,  $-\text{CF}_2\text{CH}_2\text{CF}_3$ ,  $-\text{CF}_2\text{CHFBr}$ ,  $-\text{OCF}_3$ ,  $-\text{OCF}_2\text{CHF}_2$ ,  $-\text{OCF}_2\text{CHFCl}$ ,  $-\text{OCF}_2\text{CHCl}_2$ ,  $-\text{OCF}_2\text{CHFBr}$ , and  $-\text{OCF}_2\text{CHF-CF}_3$  radicals.

Suitable heterocyclic radicals for the  $\text{R}_6\text{-N-R}_4$  or  $\text{R}_8\text{-N-R}_9$  group are, in particular, piperidine, pyrrolidine, morpholine and 2,6-dimethylmorpholine.

30

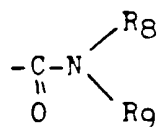
Preferred compounds of the formula I are those compounds in which

Ar denotes phenyl, which may be monosubstituted to trisubstituted by fluorine, chlorine, bromine, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>4</sub>)alkenyloxy, (C<sub>3</sub>-C<sub>4</sub>)alkynyloxy, halo(C<sub>1</sub>-C<sub>4</sub>)alkyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxy, -NHR<sub>1</sub>, (C<sub>1</sub>-C<sub>4</sub>)alkylthio, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfonyl, NO<sub>2</sub> or a radical of the formula



R<sub>1</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl,

R<sub>2</sub> and R<sub>3</sub>, independently of one another, denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, -CN, carboxyl or



R<sub>4</sub> denotes (C<sub>1</sub>-C<sub>4</sub>)alkyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxyalkyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl(C<sub>1</sub>-C<sub>4</sub>)alkyl;

R<sub>5</sub> denotes H or (C<sub>1</sub>-C<sub>4</sub>)alkyl,

R<sub>7</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl,  $\begin{array}{c} \text{R}_8 \\ \diagup \\ -\text{C}-\text{N} \\ \parallel \quad \diagdown \\ \text{X} \quad \text{R}_9 \end{array} ;$

phenyl or benzyl, which may both be monosubstituted or disubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, halogen, NO<sub>2</sub> or -CF<sub>3</sub>,

R<sub>8</sub> and R<sub>9</sub> denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or, together with the nitrogen atom connecting them, a 6-membered saturated heterocyclic ring,

M denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{C} \\ \diagdown \\ \text{R}_3 \end{array}$ , -O- or -NR<sub>7</sub>-

Q denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{C} \\ \diagdown \\ \text{R}_3 \end{array}$

T denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{C} \\ \diagdown \\ \text{R}_3 \end{array}$  or -S-

X denotes O or S,

5 Y denotes O or NH,

Z denotes O or S and

m denotes 1.

Particularly preferred compounds of the formula I are those compounds in which

- 10 Ar denotes phenyl, which may be monosubstituted to trisubstituted by fluorine, chlorine, bromine, (C<sub>1</sub>-C<sub>4</sub>)-alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>4</sub>)-alkenyloxy, (C<sub>3</sub>-C<sub>4</sub>)alkynyloxy or (C<sub>1</sub>-C<sub>4</sub>)alkylthio,
- 15 R<sub>1</sub> denotes hydrogen or (C<sub>1</sub>-C<sub>4</sub>)alkyl,

R<sub>2</sub> and R<sub>3</sub>, independently of one another, denote hydrogen or (C<sub>1</sub>-C<sub>4</sub>)alkyl,

M denotes  $\text{>CR}_2\text{R}_3$  or oxygen,

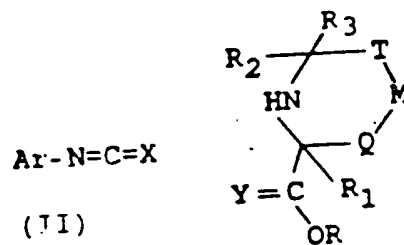
Q and T denote  $\text{>CR}_2\text{R}_3$ , and

20 X and Y denote oxygen.

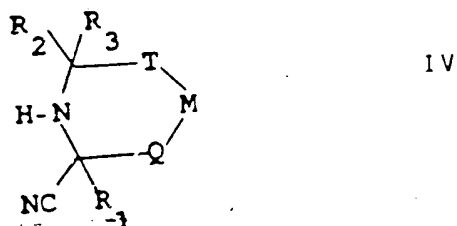
The invention furthermore relates to a process for the preparation of compounds of the formula (I), or salts

thereof, wherein

- a) where Y = O or S, a compound of the formula II is reacted with a compound of the formula III



- 5 in which R denotes H or (C<sub>1</sub>-C<sub>4</sub>)alkyl, or  
 b) a compound of the formula II is reacted with an amine of the formula IV



- or  
 10 c) where Y = O, a compound of the formula I obtained under b) is hydrolyzed, and the compound obtained is converted, if appropriate, into its salt.

In process variant a), the reaction for R = alkyl takes place in an inert organic solvent, for example an aromatic solvent such as toluene or chlorobenzene, a halogenated hydrocarbon such as chloroform, an ether such as diisopropyl ether or in dimethylformamide, if appropriate with base catalysis, at temperatures from 20 to 120°C, preferably 60 to 100°C. The bases employed are preferably organic bases, for example organic amines such as triethylamine or alternatively pyridine.

The reaction for R = hydrogen can alternatively be carried out in water as the solvent or, preferably, in the water/organic solvent 2-phase system. The procedure



is particularly preferred here in which the compound of the formula III is converted into the anion using an inorganic base, for example an alkali metal hydroxide, carbonate or hydrogen carbonate or alkaline-earth metal hydroxide, carbonate or hydrogen carbonate, such as sodium hydroxide or alternatively potassium carbonate, or an organic base, for example an organic amine such as triethylamine. In order to dissolve the anion in water, the isocyanate or isothiocyanate of the formula II, dissolved in an inert organic solvent such as, for example, toluene, chlorobenzene or chloroform is then added dropwise with vigorous stirring.

The pH of the aqueous phase is then adjusted to a value between 1 and 3 using an acid, preferably using a mineral acid such as hydrochloric acid or sulfuric acid, and subsequently reacted further at temperatures between 50 and 100°C.

In process variant b), the reaction takes place in an inert organic solvent, for example an aromatic solvent such as toluene or chlorobenzene, a halogenated hydrocarbon such as chloroform or in dimethylformamide, at temperatures from 20 to 120°C, preferably 60 to 100°C.

The hydrolysis in process variant c) takes place in water or aqueous mineral acid, if appropriate in the presence of an inert organic solvent, at temperatures between 20 and 120°C, preferably 60 and 100°C. Possible organic solvents are, for example, water-immiscible solvents such as aromatic solvents (for example toluene or chlorobenzene) or halogenated hydrocarbons (for example chloroform).

The compounds of the formula (II) are known or can be prepared analogously to known processes, cf. Houben-Weyl, Methoden der organischen Chemie [Methods of Organic Chemistry], Vol. VIII, p. 120 (1952), Houben-Weyl Vol. IX, p. 875, 869 (1955).

Some amines of the formula III are known. For Q, M and T = CR<sub>2</sub>R<sub>3</sub> and Q and T = CR<sub>2</sub>R<sub>3</sub> and M = NR<sub>7</sub>, they can be obtained by simple catalytic hydrogenation of the corresponding pyridine or pyrazine derivatives. Amines of the  
5 formula III can also be prepared from amines of the formula IV by conversion of the nitrile group by conventional methods, cf., for example, Org. Synth. Coll. Vol. I, p. 321 (1941) or Houben-Weyl Vol. VIII, p. 536.

Some amines of the general formula IV are known from JP-A  
10 3073-569 or they can be prepared analogously to the process described therein.

The compounds of the formula I according to the invention have an excellent herbicidal activity against a broad range of economically important monocotyledon and dicotyledon harmful plants. Even perennial weeds which are  
15 difficult to control and put out rhizomes, rootstocks or other permanent organs are well covered by the active compounds. At the same time, it is immaterial whether the substances are applied using presowing, preemergence  
20 or postemergence methods.

The following are examples of harmful plants which can be controlled: weed grasses, such as Avena fatua, Alopecurus sp., Lolium sp., Setaria sp., Digitaria sp., Sorghum halepense, Echinochloa sp., Agropyron sp., Cynodon sp. and  
25 Phalaris sp., and dicotyledon plants, such as Lamium sp., Veronica sp., Galium sp., Stellaria sp., Matricaria sp., Papaver sp., Centaurea sp., Amaranthus sp., Galinsoga sp., Mercurialis sp., Sida sp., Abutilon sp., Ambrosia sp., Xanthium sp., Cirsium sp., Artemisia sp., Rumex sp.,  
30 Convolvulus sp., Ipomea sp. and Sinapis sp..

If the compounds according to the invention are applied to the soil surface before germination, either emergence of the weed seedlings is entirely prevented, or the weeds grow to the seed leaf stage, but then stop growing and finally die completely after three to five weeks have

passed. On application of the active compounds to the green parts of the plants using the postemergence method, a drastic cessation of growth likewise occurs very rapidly after the treatment, and the weed plants remain  
5 at the stage of growth present at the time of application or die completely more or less quickly after a certain time, so that weed competition which is harmful for the crop plants can in this way be eliminated very early and lastingly through use of the novel agents according to  
10 the invention.

Although the compounds according to the invention have an excellent herbicidal activity against monocotyledon and dicotyledon weeds, crop plants of economically important crops, such as, for example, wheat, barley, rye, rice,  
15 corn, sugarbeet, cotton and soybean, are damaged only insignificantly or not at all. For these reasons, the present compounds are very highly suitable for selective control of undesired plant growth in agricultural crop plants.

20 In addition, the compounds according to the invention have growth-regulating properties in crop plants. They have a regulating action on the plant's inherent metabolism, and can thus be employed for simplifying harvesting, such as, for example, by initiating desiccation,  
25 abscission and stunting of growth. In addition, they are also suitable for general control and inhibition of undesired vegetative growth without at the same time killing the plants. Inhibition of vegetative growth plays an important part in many monocotyledon and dicotyledon  
30 crops since lodging can thereby be reduced or completely prevented.

The compounds according to the invention can be used in the conventional preparations, if appropriate mixed with further active compounds, as wettable powders, emulsifiable concentrates, sprayable solutions, dressings, dusts,  
35 dispersions, granules or microgranules.

Wettable powders are preparations, uniformly dispersible in water, which, besides the active compounds and in addition to a diluent or inert material, contain wetting agents, for example polyoxethylated alkylphenols, polyoxethylated fatty alcohols, alkyl- or alkylphenylsulfonates and/or dispersing agents, such as sodium ligninsulfonate, sodium 2,2'-dinaphthylmethane-6,6'-disulfonate, sodium dibutylnaphthalenesulfonate, or alternatively sodium oleoylmethyltaurinate. They are produced in a conventional fashion, for example by grinding and mixing the components.

Emulsifiable concentrates can be produced, for example, by dissolving the active compounds in an inert organic solvent, such as butanol, cyclohexanone, dimethylformamide, xylene or alternatively higher-boiling aromatics or aliphatic or cycloaliphatic hydrocarbons, with addition of one or more emulsifiers. In the case of liquid active compounds, the solvent component can also be omitted entirely or partly. As emulsifiers, the following can be used, for example: calcium salts of alkylarylsulfonic acid, such as Ca dodecylbenzenesulfonate, or nonionic emulsifiers, such as fatty acid polyglycol esters, alkylaryl polyglycol ethers, fatty alcohol polyglycol ethers, propylene oxide/ethylene oxide condensation products, fatty alcohol/propylene oxide condensation products, alkyl polyglycol ethers, sorbitan fatty acid esters, polyoxethylene sorbitan acid esters or polyoxethylene sorbitol esters.

Dusts can be obtained by grinding the active compounds with finely divided solids, for example talc, natural clays, such as kaolin, bentonite, pyrophyllite or diatomaceous earth.

Granules can be produced either by atomizing the active compounds onto adsorptive, granulated inert material or by applying active compound concentrates onto the surface of carrier materials, such as sand or kaolinite, or of

granulated inert material by means of binders, for example polyvinyl alcohol, sodium polyacrylate or alternatively mineral oils. Suitable active compounds can also be granulated in the manner which is conventional for the production of fertilizer granules - if desired as a mixture with fertilizers.

The active compound concentration in wettable powders is about 10 to 90% by weight, the remainder to 100% by weight comprising conventional formulation components. In the case of emulsifiable concentrates, the active compound concentration can be about 5 to 80% by weight. Dust formulations usually contain 0.05 to 20% by weight of active compound(s), and sprayable solutions about 2 to 20% by weight. In the case of granules, the active compound content depends partly on whether the active compound is present in liquid or solid form and on which granulation auxiliaries, fillers etc. are used.

In addition, the active compound formulations mentioned contain, if appropriate, the adhesives, wetting agents, dispersants, emulsifiers, penetrants, solvents, fillers or excipients which are conventional in each case.

For use, the concentrates, present in commercially available form, are diluted, if appropriate, in a conventional fashion, for example using water in the case of wettable powders, emulsifiable concentrates, dispersions and, in some cases, also in the case of microgranules. Dusts and granulated formulations, and also sprayable solutions, are usually not further diluted with further inert substances before use.

The application rate necessary varies with the external conditions, such as temperature, humidity, inter alia. It can vary within broad limits, for example between 0.005 and 10.0 kg/ha or more of active substance, but is preferably between 0.01 and 2 kg/ha.

Mixtures of the mixed formulations with other active compounds, such as, for example, insecticides, acaricides, herbicides, fertilizers, growth regulators or fungicides, may also be possible.

5 The invention is illustrated by the following examples.

A. Formulation examples

A dust is obtained by mixing 10 parts by weight of active compound and 90 parts by weight of talc or inert material, and comminuting the mixture in a hammer mill.

- 10 A wettable powder which is easily dispersible in water is obtained by mixing 25 parts by weight of active compound, 64 parts by weight of kaolin-containing quartz as inert material, 10 parts by weight of potassium lignin-sulfonate and 1 part by weight of sodium oleoylemethyl-  
15 taurinate as wetting and dispersing agent, and grinding the mixture in a pin disk mill.

- A dispersing concentrate which is easily dispersible in water is obtained by mixing 20 parts by weight of active compound with 6 parts by weight of alkylphenol polyglycol  
20 ether (Triton X 207), 3 parts by weight of isotridecanol polyglycol ether (8 E0) and 71 parts by weight of paraffinic mineral oil (boiling range, for example, about 255 to above 377°C), and grinding the mixture in a ball mill to a fineness below 5 microns.

- 25 An emulsifiable concentrate is obtained from 15 parts by weight of active compound, 75 parts by weight of cyclohexanone as solvent and 10 parts by weight of oxethylated nonylphenol (10 E0) as emulsifier.

Chemical Examples

Example 1

1-Ethoxycarbonylethyl 8-(4-chloro-3-ethoxycarbonylphenyl)-7,9-dioxo-1,8-diazabicyclo[4.3.0]nonane-2-carboxylate

- 5 37.3 g (0.10 mol) of bis-(1-ethoxycarbonylethyl) piperidine-2,6-biscarboxylate were dissolved in 200 ml of toluene. 22.6 g (0.10 mol) of 4-chloro-3-ethoxycarbonylphenyl isocyanate, dissolved in 50 ml of toluene, were added dropwise at 20 - 30°C. After stirring for 1 hour at room temperature, the mixture was stirred for a further 3 hours at 80°C. After removing the solvent by distillation, the solid which remained was recrystallized from methanol. 39.5 g (82% of theory) of 1-ethoxycarbonylethyl 8-(4-chloro-3-ethoxycarbonylphenyl)-7,9-dioxo-1,8-diazabicyclo[4.3.0]nonane-2-carboxylate were obtained in the form of colorless crystals of melting point: 121 - 128°C.

Example 2

20 8-(4-Chloro-2-fluoro-5-methoxyphenyl)-2-methyl-7,9-dioxo-1,8-diazabicyclo[4.3.0]nonane

- 20.2 g (0.10 mol) of 4-chloro-2-fluoro-5-methoxyphenyl isocyanate, dissolved in 100 ml of toluene, were added dropwise to 17.1 g (0.10 mol) of ethyl 6-methylpiperidine-2-carboxylate in 100 ml of toluene. The mixture was stirred for 3 hours at 100°C. The solvent was removed by distillation under reduced pressure, and the residue was dried in a high vacuum to constant weight. 33.0 g (99% of theory) of 8-(4-chloro-2-fluoro-5-methoxyphenyl)-2-methyl-7,9-dioxo-1,8-diazabicyclo[4.3.0]nonane were obtained in the form of a pale yellow syrup.

Example 3

Ethyl 8-(4-chloro-2-fluoro-5-methoxyphenyl)-7,9-dioxo-1,8-diazabicyclo[4.3.0]nonane-3-carboxylate

20.2 g (0.1 mol) of 4-chloro-2-fluoro-5-methoxyphenyl isocyanate, dissolved in 100 ml of chlorobenzene, were added dropwise to 22.9 g (0.1 mol) of diethyl piperidine-2,5-dicarboxylate in 150 ml of chlorobenzene. The mixture was stirred for 3 hours at 100°C, the solvent was removed by distillation, and the residue was dried in a high vacuum to constant weight. 37.8 g (98% of theory) of ethyl 8-(4-chloro-2-fluoro-5-methoxyphenyl)-7,9-dioxo-1,8-diazabicyclo[4.3.0]nonane-3-carboxylate were obtained in the form of a honey-colored syrup.

Example 4

Ethyl 8-(4-chloro-2-fluoro-5-methoxyphenyl)-7,9-dioxo-1,4,8-triazabicyclo[4.3.0]nonane-5-carboxylate

10.1 g (0.05 mol) of 4-chloro-2-fluoro-5-methoxyphenyl isocyanate, dissolved in 70 ml of toluene, were added dropwise within 1 hour to 11.5 g (0.05 mol) of diethyl piperazine-2,3-dicarboxylate in 100 ml of toluene at 20°C. The mixture was subsequently stirred for 4 hours at 100°C, and the solvent was removed by distillation. The residue which remained was triturated with hexane and the solid was filtered off under suction. 14.8 g (77% of theory) of ethyl 8-(4-chloro-2-fluoro-5-methoxyphenyl)-7,9-dioxo-1,4,8-triazabicyclo[4.3.0]nonane-5-carboxylate were obtained in the form of pale yellow crystals of melting point 56 - 58°C.

Example 5

8-(4-Chlorophenyl)-7-imino-9-oxo-1,8-diazabicyclo[4.3.0]nonane

15.4 g (0.1 mol) of 4-chlorophenyl isocyanate, dissolved in 50 ml of toluene, were added dropwise to 11.0 g (0.1 mol) of 2-cyanopiperidine in 80 ml of toluene. The



mixture was stirred for 4 hours at 100°C, and the solvent was removed by distillation. 26.0 g (98% of theory) of 8-(4-chlorophenyl)-7-imino-9-oxo-1,8-diazabicyclo[4.3.0]nonane were obtained in the form of a pale brown glassy material.

Example 6

8-(4-chloro-2-fluoro-5-methoxyphenyl)-3,5-dimethyl-7,9-dioxo-4-oxa-1,8-diazabicyclo[4.3.0]nonane

20.2 g (0.10 mol) of 4-chloro-2-fluoro-5-methoxyphenyl isocyanate, dissolved in 100 ml of toluene, were added dropwise to 13.9 g (0.10 mol) of 3-cyano-2,6-dimethylmorpholine in 100 ml of toluene. The mixture was stirred for 3 hours at 100°C. After 50 ml of 20% strength aqueous hydrochloric acid had been added, the mixture was stirred for a further 2 hours at 100°C. The organic phase was separated off, washed twice with 100 ml of water and dried over sodium sulfate. The tacky, solid residue remaining after the solvent had been stripped off was triturated with diisopropyl ether. 26.4 g (77% of theory) of 8-(4-chloro-2-fluoro-5-methoxyphenyl)-3,5-dimethyl-7,9-dioxo-4-oxa-1,8-diazabicyclo[4.3.0]nonane were obtained in the form of colorless crystals of melting point 126 - 131°C.

Example 7

8-(4-chloro-2-fluoro-5-methoxyphenyl)-7,9-dioxo-3-thia-1,8-diazabicyclo[4.3.0]nonane

20.2 g (0.10 mol) of 4-chloro-2-fluoro-5-methoxyphenyl isocyanate, dissolved in 50 ml of toluene, were added dropwise to 17.5 g (0.10 mol) of ethyl 1,3-thiazane-4-carboxylate in 150 ml of toluene. The mixture was stirred for 4 hours at 100°C, and the solvent was removed by distillation. The solid which remained was triturated with di-isopropyl ether and dried. 24.9 g (75% of theory) of 8-(4-chloro-2-fluoro-5-methoxyphenyl)-7,9-dioxo-3-thia-1,8-diazabicyclo[4.3.0]nonane were

obtained in the form of pale yellow crystals of melting point 55 - 61°C.

Example 8

5 8-(4-Chloro-2-fluoro-5-propargyloxyphenyl)-2-(1,3-dioxo-  
lan-2-yl)-7,9-dioxo-1,8-diazabicyclo[4.3.0]nonane

11.3 g (0.05 mol) of 4-chloro-2-fluoro-5-propargyloxy-  
phenyl isocyanate, dissolved in 50 ml of toluene, were  
added dropwise to 11.5 g (0.05 mol) of ethyl 6-(1,3-  
dioxolan-2-yl)-piperidine-2-carboxylate in 100 ml of  
10 toluene. After the mixture had been stirred for 3 hours  
at 100°C, the solvent was removed by distillation, and  
the residue was dried in a high vacuum to constant weight.  
19.8 g (97% of theory) of 8-(4-chloro-2-fluoro-5-propargyl-  
oxyphenyl)-2-(1,3-dioxolan-2-yl)-7,9-dioxo-1,8-diaza-  
15 bicyclo[4.3.0]nonane were obtained in the form of a  
colorless glass.

Example 9

4-Benzyl-8-(4-chloro-2-fluoro-5-isopropoxyphenyl)-7,9-  
dioxo-1,4,8-triazabicyclo[4.3.0]nonane

20 11.5 g (0.05 mol) of 4-chloro-2-fluoro-5-isopropoxyphenyl  
isocyanate, dissolved in 50 ml of toluene, were added  
dropwise to 12.4 g (0.05 mol) of ethyl 4-benzylpiperazine-  
2-carboxylate in 100 ml of toluene. The mixture was  
stirred for 3 hours at 100°C, and the solvent was removed  
25 by distillation. After drying the residue in HV<sup>1)</sup> to  
constant weight, 30.7 g (96% of theory) of 4-benzyl-8-(4-  
chloro-2-fluoro-5-isopropoxyphenyl)-7,9-dioxo-1,4,8-  
triazabicyclo[4.3.0]nonane were obtained in the form of a  
pale yellow syrup.

30 HV = high vacuum


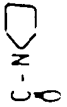
Example 10

2,2,2-Trifluoroethyl 8-(4-chloro-2-fluoro-5-methoxy-phenyl)-7-oxo-9-thioxo-1,8-diazabicyclo[4.3.0]nonane-2-carboxylate

- 5 10.9 g (0.05 mol) of 4-chloro-2-fluoro-5-methoxyphenyl isothiocyanate, dissolved in 70 ml of toluene, were added dropwise to 16.9 g (0.05 mol) of bis(2,2,2-trifluoroethyl) piperidine-2,6-dicarboxylate in 150 ml of toluene, and the mixture was stirred for 6 hours at 100°C. After the  
10 solvent had been removed by distillation, the residue was triturated with n-hexane and dried. 19.8 g (87% of theory) of 2,2,2-trifluoroethyl 8-(4-chloro-2-fluoro-5-methoxyphenyl)-7-oxo-9-thioxo-1,8-diazabicyclo[4.3.0]nonane-2-carboxylate were obtained in the form of pale  
15 beige crystals of melting point 134 - 138°C.

The compounds (I) of Table 1 below can be prepared in an analogous fashion.

Table 1

Example	R <sub>1</sub>	X	Y	Q	M	T	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	M.P. (°C.)
11	4-Cl-C <sub>6</sub> H <sub>4</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H		-H	Glass
12	4-Cl-2-F-5-OCCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-H	Syrup
13	4-Cl-2-F-5-OCCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CO <sub>2</sub> CH <sub>2</sub> CF <sub>3</sub>	-H	104-109
14	4-Cl-2-F-5-OCCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-C-N< 	-H	Glass
15	"	0	0	-CH <sub>2</sub> -	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-CH <sub>2</sub> -	-H	-H	-H	Syrup
16	"	0	0	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	Resin
17	"	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CO <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>	-H	Glass
18	"	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CO <sub>2</sub> CH <sub>2</sub> -OCH <sub>3</sub>	-H	Glass
19	"	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CO <sub>2</sub> CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>   CH <sub>3</sub>	-H	Glass
20	"	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CH <sub>2</sub> OH	-H	Resin
21	4-Cl-2-F-5-OCCH(CH <sub>3</sub> ) <sub>2</sub> - -C <sub>6</sub> H <sub>2</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH-C <sub>2</sub> H <sub>5</sub>	-H	-H	-H	Syrup

Example	Ar	X	Y	Q	M	T	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	M.P.(°C)
22	4-Cl-2F-5- $\text{OCH}_3$ - $\text{C}_6\text{H}_2$	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -H, H	-H	-H	-H	Syrup
23	"	0	0	-CH <sub>2</sub> -	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-CH <sub>2</sub> -H-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-H	-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-H	Glass
24	2,4-F <sub>2</sub> -5-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>2</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-CH <sub>3</sub>	-H	-CH <sub>3</sub>	-H	Syrup
25	4-Br-3-CO <sub>2</sub> -CH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>3</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-CH <sub>3</sub>	-H	-CH <sub>3</sub>	-H	Syrup
26	4-Cl-3-CO <sub>2</sub> CH(CH <sub>3</sub> )CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>2</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH-CO <sub>2</sub> CH <sub>3</sub> -H-CH <sub>3</sub>	-H	-H	-H	Glass
27	4-Cl-3-CO <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>	0	0	-CH-CO <sub>2</sub> CH <sub>3</sub>	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-H	-H	-H	-H	Glass
28	4-Cl-2-F-5-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>2</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-CO <sub>2</sub> CH <sub>3</sub>	-H	-CO <sub>2</sub> CH <sub>3</sub>	-H	Syrup
29	3-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -4-OC <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-CH <sub>3</sub>	-H	-CH <sub>3</sub>	-H	Syrup
30	3-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -4-OC <sub>2</sub> H <sub>5</sub> CF <sub>3</sub> -C <sub>6</sub> H <sub>3</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-CO <sub>2</sub> CH <sub>2</sub> CF <sub>3</sub>	-H	-CO <sub>2</sub> CH <sub>2</sub> CF <sub>3</sub>	-H	Glass
31	2-F-4-OC <sub>2</sub> H <sub>5</sub> -5-OC <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>2</sub>	0	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-CH <sub>3</sub>	-H	-CH <sub>3</sub>	-H	Oil
32	4-Cl-2-F-5-OC <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>2</sub>	5	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-CH <sub>3</sub>	-H	-CH <sub>3</sub>	-H	Resin
33	"	5	0	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH-C <sub>2</sub> H <sub>5</sub> -H-H	-H	-H	-H	Resin
34	"	5	0	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-CH <sub>2</sub> -	-CH <sub>2</sub> -H-H	-H	-H	-H	Resin
35	"	5	0	-CH <sub>2</sub> -	-CH-CO <sub>2</sub> CH <sub>3</sub>	-CH <sub>2</sub> -H-H	-H	-H	-H	Glass

Example	Ar	$\lambda$	Y	Q	M	T	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	M.p.(°C)
36	4-Cl-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	S	C	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CO <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>	-H	Glass
37	"	S	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CO <sub>2</sub> CH <sub>2</sub> CO <sub>2</sub> CH <sub>3</sub>	-H	Syrup
38	4-Br-3-CO <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>3</sub>	S	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-CH <sub>3</sub>	-H	Resin
39	4-Cl-3-CO <sub>2</sub> CH(CH <sub>3</sub> )CO <sub>2</sub> CH <sub>3</sub> -C <sub>6</sub> H <sub>3</sub>	S	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	Syrup
40	4-Cl-2,5-OCH <sub>2</sub> -CO <sub>2</sub> CH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	O	O	-CH-CO <sub>2</sub> CH <sub>3</sub>	-NH-	-CH <sub>2</sub> -	-H	-H	-H	Resin
41	4-Cl-2-F-5-CO <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>2</sub>	O	O	-CH <sub>2</sub> -	-N-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin
42	4-Cl-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	O	O	-CH <sub>2</sub> -	-N-C(=O)CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Glass
43	4-Cl-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	O	O	-CH-CO <sub>2</sub> CH <sub>3</sub>	-N-C(=O)CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Glass
44	"	O	O	-CH <sub>2</sub> -	-N-CO <sub>2</sub> CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Syrup
45	"	O	O	-CH <sub>2</sub> -	-N-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin
46	"	O	O	-CH-CO <sub>2</sub> CH <sub>3</sub>	-N-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin
47	"	O	O	-CH-CO <sub>2</sub> CH <sub>3</sub>	-N-CO <sub>2</sub> CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin

Sample	Ar	X	Y	Q	M	T	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	M.P.(°C)
48	4-Br-5-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	0	0	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-NH	-OL <sub>2</sub> -	-H	-H	-H	Resin
49	4-Cl-5-CO <sub>2</sub> CH(CH <sub>3</sub> )CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	0	0	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-N-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin
50	4-Cl-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	S	0	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-NH	-CH <sub>2</sub> -	-H	-H	-H	Glass
51	"	S	0	-CH <sub>2</sub> -	-H-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Syrup
52	"	S	0	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-N-CO <sub>2</sub> CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin
53	"	S	0	-CH <sub>2</sub> -	-N-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin
54	"	0	0	-CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	-H	-H	-H	Syrup
55	4-Cl-C <sub>6</sub> H <sub>4</sub>	0	0	-CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	-H	-H	-H	121-125
56	4-Br-3-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	0	0	-CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	-H	-H	-H	Syrup
57	4-Cl-3-CO <sub>2</sub> CH(CH <sub>3</sub> )CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	0	0	-OL <sub>2</sub> -	-O-	-OL <sub>2</sub> -	-H	-H	-H	Resin
58	4-Cl-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	S	0	-CH <sub>2</sub>	-O-	-CH <sub>2</sub>	-H	-H	-H	97-102
59	"	S	0	-CH-CH <sub>3</sub>	-O-	-CH-CH <sub>3</sub> -H	-H	-H	-H	Glass
60	4-Cl-C <sub>6</sub> H <sub>4</sub>	0	0	-CH <sub>2</sub> -	-S-	-CH <sub>2</sub> -	-H	-CH <sub>3</sub>	-H	Syrup

Example	Ar	X	Y	Q	M	T	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	M.p.(°C)
61	4-Cl-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	0	0	-CH-CH <sub>3</sub>	-S-	-CH-CH <sub>3</sub>	-H	-H	-H	Resin
62	"	S	0	-CH-CH <sub>3</sub>	-S-	-CH-CH <sub>3</sub>	-H	-H	-H	Resin
63	"	0	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	48-53
64	4-Br-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	0	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	75-80
65	4-Cl-3-CO <sub>2</sub> CH(CH <sub>3</sub> )CO <sub>2</sub> C <sub>6</sub> H <sub>3</sub>	0	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	Syrup
66	3-5-Cl <sub>2</sub> -4-CF <sub>3</sub> CHFCF <sub>2</sub> -C <sub>6</sub> H <sub>2</sub>	0	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	102-106
67	4-1-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	0	NH	-CH <sub>2</sub> -	-CH-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Glass
68	"	0	NH	-CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	-H	-H	-H	142-147
69	4-Cl-2-F-5-OCH <sub>2</sub> C≡CH-C <sub>6</sub> H <sub>2</sub>	0	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	112-116
70	4-Cl-2-F-5-OCH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>2</sub>	0	NH	-CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	-H	-H	-H	Glass
71	4-Br-3-CO <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>3</sub>	0	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	86-90
72	4-Cl-2-F-5-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>2</sub>	0	NH	-CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	-H	-H	-H	Glass
73	4-OCH <sub>3</sub> -3-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	0	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	Resin
74	4-SC <sub>2</sub> H <sub>5</sub> -3-(O <sub>2</sub> CH(CH <sub>3</sub> )) <sub>2</sub> -C <sub>6</sub> H <sub>3</sub>	0	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	-H	Resin



Example	Ar	X	Y	Q	M	T	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	m.p.(°C)
75	4-OOCH <sub>2</sub> α <sub>3</sub> -3-OO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	O	NH	CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	-H	-H	-H	Resin
76	4-Cl-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>2</sub>	O	NH	-CH <sub>2</sub> -	-CH-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Glass
77	"	O	NH	-CH-CH <sub>3</sub>	-CH <sub>2</sub> -	-CH-CH <sub>3</sub>	-H	-H	-H	Glass
78	"	O	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	CH <sub>3</sub>	-CH <sub>3</sub>	-H	Syrup
79	"	O	NH	-CH <sub>2</sub> -	-CH-CH <sub>3</sub>	-CH <sub>2</sub> -	CH <sub>3</sub>	-CH <sub>3</sub>	-H	Syrup
80	"	O	NH	-CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	CH <sub>3</sub>	-CH <sub>3</sub>	-H	Resin
81	"	O	NH	-CH <sub>2</sub> -	-NH-	-CH <sub>2</sub> -	-H	-H	-H	Resin
82	"	O	NH	-CH <sub>2</sub> -	-N-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Syrup
83	"	O	NH	-CH <sub>2</sub> -	-N-C <sub>6</sub> H <sub>5</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin
84	"	O	NH	-CH <sub>2</sub> -	-N-C-NCH <sub>3</sub> 0	-CH <sub>2</sub> -	-H	-H	-H	Resin
85	"	O	NH	-CH <sub>2</sub> -	-N-P=O γOC <sub>2</sub> H <sub>5</sub> ) <sub>2</sub>	-CH <sub>2</sub> -	-H	-H	-H	Glass
86	"	S	NH	-CH <sub>2</sub> -	-CH-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Glass
87	4-Cl-2-F-5-OOCH <sub>2</sub> C≡CH-C <sub>6</sub> H <sub>2</sub>	S	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-H	-H	H	Glass
88	4-Cl-2-F-5-OCH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>2</sub>	S	NH	-CH-CH <sub>3</sub>	-CH <sub>2</sub> -	-CH-CH <sub>3</sub>	-H	-H	-H	Resin

Example	Ar	X	Y	Q	M	T	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	M.p.(°C.)
89	4-Cl-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>5</sub>	S	NH	-CH <sub>2</sub> -	-O-	-CH <sub>2</sub> -	-H	-H	-H	Glass
90	"	S	NH	-CH-CH <sub>3</sub>	-O-	-CH-CH <sub>3</sub>	-H	-H	-H	Glass
91	"	S	NH	-CH <sub>2</sub> -	-S-	-CH <sub>2</sub> -	-H	-H	-H	Syrup
92	"	S	NH	-CH <sub>2</sub> -	-CH-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub>	-CH <sub>2</sub> -	-H	-H	-H	Syrup
93	"	S	NH	-CH <sub>2</sub> -	-NH-	-CH <sub>2</sub> -	-H	H	-H	Resin
94	"	S	NH	-CH <sub>2</sub> -	-N-CH <sub>3</sub>	-CH <sub>2</sub> -	-H	-H	-H	Resin
95	"	S	NH	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin
96	"	S	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin
97	4-Br-2-F-5-OCH <sub>3</sub> -C <sub>6</sub> H <sub>5</sub>	S	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin
98	4-Br-2F-5-OCH <sub>2</sub> C≡CH-C <sub>6</sub> H <sub>2</sub>	O	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin
99	4-Cl-2F-5OCH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>2</sub>	O	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin
100	4-Cl-2F-5-OCH <sub>2</sub> C≡CH-C <sub>6</sub> H <sub>2</sub>	S	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Syrup
101	4-Br-5-CO <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>3</sub>	S	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin
102	4-Cl-5-CO <sub>2</sub> CH(CH <sub>3</sub> )CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	O	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin
103	4-OCH <sub>2</sub> CF <sub>3</sub> -3-CO <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	O	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin
104	2,4-F <sub>2</sub> -5-CO <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub> -C <sub>6</sub> H <sub>2</sub>	O	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Syrup
105	4-OCH <sub>2</sub> C <sub>2</sub> H <sub>5</sub> -3-Cl-C <sub>2</sub> H <sub>5</sub> -C <sub>6</sub> H <sub>3</sub>	S	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Syrup
106	4-SC <sub>2</sub> H <sub>5</sub> -3-CO <sub>2</sub> CH <sub>2</sub> CF <sub>3</sub> -C <sub>6</sub> H <sub>3</sub>	O	O	-CH <sub>2</sub> -	-CH <sub>2</sub> -	-S-	-H	-H	-H	Resin

Biological Examples

The damage to weed plants or the crop plant compatibility was assessed according to a key, in which the effectiveness is expressed by values from 0 - 5. In this:

0 = no action or damage

1 = 0 - 20% action or damage

2 = 20 - 40% action or damage

3 = 40 - 60% action or damage

4 = 60 - 80% action or damage

5 = 80 - 100% action or damage

1. Pre-emergent action on weeds

Seeds or rhizome pieces of monocotyledonous and dicotyledonous weed plants were placed in sandy loam in plastic pots ( $\phi$  = 9 cm) and covered with soil. The compounds according to the invention, formulated in the form of wettable powders or emulsion concentrates, were then applied in varying dosages to the surface of the covering soil as aqueous suspensions or emulsions with an applicational amount of water equivalent to 600 - 800 l/ha.

After treatment, the pots were placed in a greenhouse and kept under good growth conditions for the weed plants (temperature 23 plus/minus 1°C, relative air humidity 60 - 80%).

The optical assessment of the plants or emergence damage in comparison to the untreated controls was carried out after emergence of the test plants after an experimental time of 3 - 4 weeks.

As the assessment values in Table 2 show, the compounds according to the invention have a good herbicidal pre-emergence activity against a broad spectrum of weed

grasses and weeds.

## 2. Post-emergent action against weeds

5 Seeds or rhizome pieces of monocotyledonous and dicotyledonous weeds were placed in sandy loam in plastic pots ( $\phi = 9$  cm), covered with soil, and germinated in a greenhouse under good growth conditions. The test plants were treated at the three-leaf stage three weeks after sowing.

10 The compounds according to the invention, formulated as wettable powders or as emulsion concentrates, were sprayed onto the green plant parts in various dosages having an applicational amount of water equivalent to 600 l/ha, and the action of the preparations was assessed optically in comparison to untreated controls after the test plants had stood for about 3 - 4 weeks in a greenhouse under optimum growth conditions (temperature 23 plus/minus 1°C, relative air humidity 60 - 80%).

20 The agents according to the invention also have a good post-emergent herbicidal activity against a broad spectrum of economically important weed grasses and weeds (Table 3).

Tabelle 2: Pre-emergent action of the compounds

Example No.	Dose kg of a.i./ha	herbicidal action				
		STM	CRS	SIA	LOM	ECG
2	2.5	5	5	5	5	5
5	2.5	3	4	2	2	4
6	2.5	5	5	5	5	5
7	2.5	5	5	5	5	5
54	2.5	5	5	5	5	5
55	2.5	5	5	5	5	5
63	2.5	5	5	5	5	5
64	2.5	5	5	5	5	5
67	2.5	5	5	5	5	5
68	2.5	5	5	5	5	5

Tabelle 3: Post-emergent action of the compounds

Example	Dose kg of a.i./ha	herbicial action				
		STM	CRS	SIA	LOM	ECG
2	2.5	5	5	5	5	5
6	2.5	5	4	5	5	5
7	2.5	5	4	5	5	4
54	2.5	5	5	5	5	5
55	2.5	4	3	3	3	4
63	2.5	5	4	5	5	3
64	2.5	4	3	5	4	4
67	2.5	4	5	5	3	3
68	2.5	5	5	5	5	5

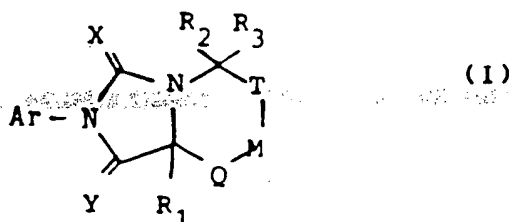
Abbreviations

STM = Stellaria media  
 CRS = Chrysanthemum segetum  
 SIA = Sinapis alba  
 LOM = Lolium multiflorum  
 ECG = Echinochloa crus-galli  
 a.i. = active substance

XXXXXXXXXXXX  
PATENT CLAIMS:

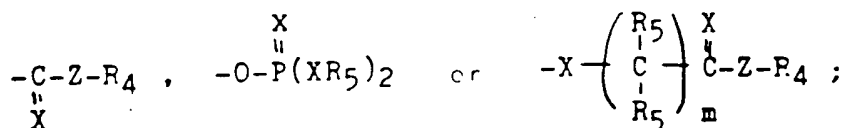
THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS: HOE 86/F 312

1. A compound of the formula I, or a salt thereof,



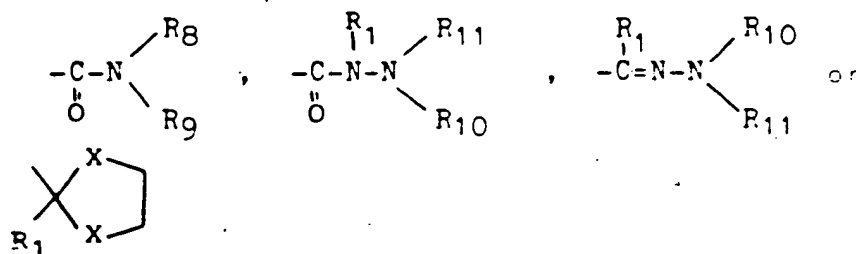
in which

Ar denotes phenyl, naphthyl, pyridyl, quinolinyl or isoquinolinyl, these radicals being monosubstituted to tetrasubstituted, preferably monosubstituted to trisubstituted, by identical or different radicals from the series comprising halogen, hydroxyl, (C<sub>1</sub>-C<sub>4</sub>)alkyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>6</sub>)cycloalkoxy, (C<sub>3</sub>-C<sub>6</sub>)alkenyloxy, (C<sub>3</sub>-C<sub>6</sub>)alkynyloxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxy-(C<sub>1</sub>-C<sub>4</sub>)-alkoxy, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxy, halo(C<sub>3</sub>-C<sub>6</sub>)alkenyloxy, halo(C<sub>3</sub>-C<sub>6</sub>)alkynyloxy, (C<sub>1</sub>-C<sub>4</sub>)alkylthio, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfinyl, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfonyl, NO<sub>2</sub>, -CN, -NHR<sub>1</sub>, cyano(C<sub>1</sub>-C<sub>4</sub>)alkyl, phenoxy, phenoxy(C<sub>1</sub>-C<sub>4</sub>)alkyl, phenyl(C<sub>1</sub>-C<sub>4</sub>)alkoxy, phenylthio, phenylsulfinyl or phenylsulfonyl, it being possible for the six lastmentioned radicals to be monosubstituted, disubstituted or trisubstituted in the phenyl ring by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, -CN or NO<sub>2</sub>, or furthermore by a radical of the formula

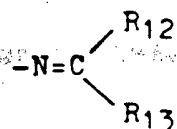


R<sub>1</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or phenyl which may be monosubstituted or disubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, NO<sub>2</sub>, CN or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl,

$R_2$  and  $R_3$ , independently of one another, denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl which may be substituted by cyano, hydroxyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxy, phenyl or benzyl which may both be substituted, in each case preferably monosubstituted or disubstituted, in the phenyl ring by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)-alkoxy, NO<sub>2</sub> or -CN; (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)-alkoxycarbonyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)-alkoxy(C<sub>1</sub>-C<sub>4</sub>)-alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)-alkoxycarbonyl(C<sub>1</sub>-C<sub>4</sub>)-alkoxycarbonyl, carboxyl or a radical of the formula



$R_4$  denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl which may be monosubstituted to hexasubstituted by halogen and/or monosubstituted or disubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxy(C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkylthio, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfinyl, (C<sub>1</sub>-C<sub>4</sub>)-alkylsulfonyl, (C<sub>1</sub>-C<sub>4</sub>)alkylamino, -CN, furyl, tetrahydrofuryl, benzofuryl, phenyl, phenoxy or benzyloxy, it being possible for the six last-mentioned radicals to be monosubstituted to trisubstituted, in each case in the phenyl ring or the heteroaromatic ring, by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>6</sub>)cycloalkyl, (C<sub>3</sub>-C<sub>6</sub>)-alkenyl, cyclohexenyl, (C<sub>3</sub>-C<sub>6</sub>)alkynyl or phenyl, which may be monosubstituted to trisubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl; or a radical of the formula

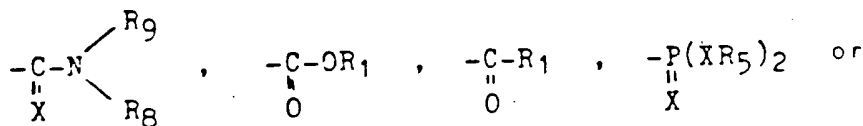


where the lastmentioned radical is excluded when  
Z = S,

R<sub>5</sub> denotes hydrogen or (C<sub>1</sub>-C<sub>4</sub>)alkyl,

R<sub>6</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or, together with  
R<sub>4</sub> and the nitrogen atom connecting these radi-  
cals, a 5- to 7-membered heterocyclic ring which  
can contain as ring members one or two radicals  
from the series comprising -O-, -S- and -NR<sub>5</sub>- and  
which may be monosubstituted to trisubstituted by  
(C<sub>1</sub>-C<sub>4</sub>)alkyl;

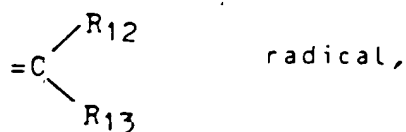
R<sub>7</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, phenyl or benzyl,  
where the phenyl ring may in each case be monosub-  
stituted to trisubstituted by halogen, (C<sub>1</sub>-C<sub>4</sub>)-  
alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl,  
-NO<sub>2</sub>, CF<sub>3</sub>, -CN or a radical of the formula





R<sub>8</sub> and R<sub>9</sub>, independently of one another, denote hydrogen or (C<sub>1</sub>-C<sub>4</sub>)alkyl, or the two radicals R<sub>8</sub> and R<sub>9</sub>, together with the nitrogen atom connecting them, denote a 5- to 7-membered heterocyclic ring which can contain as ring members one or two radicals from the series comprising -O-, -S- and -NR<sub>5</sub>- and which may be monosubstituted to trisubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkyl, hydroxyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in the phenyl ring by (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, halogen or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl,

R<sub>10</sub> and R<sub>11</sub>, independently of one another, denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in each case in the phenyl ring, by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, NO<sub>2</sub>, -CN, CF<sub>3</sub> or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, or R<sub>10</sub> and R<sub>11</sub> together denote the



R<sub>12</sub> and R<sub>13</sub>, independently of one another, denote (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, phenyl or benzyl which may both be substituted, preferably monosubstituted to trisubstituted, in the phenyl ring by halogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, CF<sub>3</sub>, -CN or NO<sub>2</sub>,

M denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{=C} \\ \diagdown \\ \text{R}_3 \end{array}$ , -S-, -S-, -S-, -O- or NR<sub>7</sub>;

Q denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{=C} \\ \diagdown \\ \text{R}_3 \end{array}$ , -S- or -O-;

T denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{C} \\ \diagdown \\ \text{R}_3 \end{array}$ ,  $-\text{S}-$ ,  $\begin{array}{c} \text{O} \\ \parallel \\ -\text{S}- \end{array}$ ,  $\begin{array}{c} \text{O} \\ \parallel \\ -\text{S}- \\ \parallel \\ \text{O} \end{array}$  or  $-\text{O}-$ ,

X denotes O or S,

Y denotes O, S or NH,

Z denotes O, S or NR<sub>6</sub>,

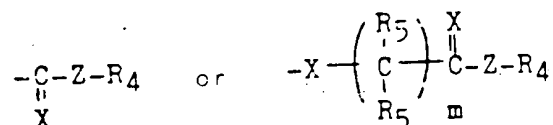
m denotes 1, 2 or 3, and

n denotes 1 or 2,

with the proviso that m cannot be CH<sub>2</sub>, S, SO or SO<sub>2</sub> when Y denotes O, R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> denote H and Q and T denotes CH<sub>2</sub>.

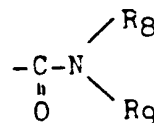
2. A compound of the formula I of claim 1, in which

Ar denotes phenyl, which may be monosubstituted to trisubstituted by fluorine, chlorine, or bromine, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, (C<sub>3</sub>-C<sub>4</sub>)alkenyloxy, (C<sub>3</sub>-C<sub>4</sub>)alkynyloxy, halo(C<sub>1</sub>-C<sub>4</sub>)alkyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxy, -NHR<sub>1</sub>, (C<sub>1</sub>-C<sub>4</sub>)alkylthio, (C<sub>1</sub>-C<sub>4</sub>)alkylsulfonyl, NO<sub>2</sub> or a radical of the formula



R<sub>1</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl,

R<sub>2</sub> and R<sub>3</sub>, independently of one another, denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl(C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl, -CN, carboxyl or



R<sub>4</sub> denotes (C<sub>1</sub>-C<sub>4</sub>)alkyl, halo(C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxyalkyl or (C<sub>1</sub>-C<sub>4</sub>)alkoxycarbonyl(C<sub>1</sub>-C<sub>4</sub>)alkyl;

R<sub>5</sub> denotes H or (C<sub>1</sub>-C<sub>4</sub>)alkyl,

R<sub>7</sub> denotes hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl,  $\begin{array}{c} \text{R}_8 \\ \diagup \\ \text{---C---N---} \\ \parallel \quad \diagdown \\ \text{X} \quad \text{R}_9 \end{array}$ , phenyl or benzyl, which may both be monosubstituted or disubstituted by (C<sub>1</sub>-C<sub>4</sub>)alkyl, (C<sub>1</sub>-C<sub>4</sub>)alkoxy, halogen, NO<sub>2</sub> or -CF<sub>3</sub>,

R<sub>8</sub> and R<sub>9</sub> denote hydrogen, (C<sub>1</sub>-C<sub>4</sub>)alkyl or, together with the nitrogen atom connecting them, a 6-membered saturated heterocyclic ring,

M denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{---C---} \\ \diagdown \\ \text{R}_3 \end{array}$ , -O- or -NR<sub>7</sub>-

Q denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{---C---} \\ \diagdown \\ \text{R}_3 \end{array}$

T denotes  $\begin{array}{c} \text{R}_2 \\ \diagup \\ \text{---C---} \\ \diagdown \\ \text{R}_3 \end{array}$  or -S-

X denotes O or S,

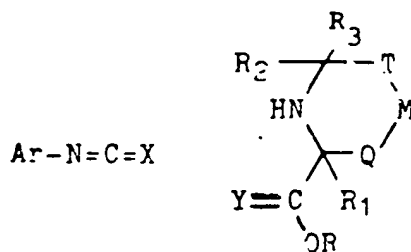
Y denotes O or NH,

Z denotes O or S and

n denotes 1.

3. A process for the preparation of a compound of the formula I of claim 1 or 2, or a salt thereof, wherein

a) where Y = O or S, a compound of the formula II is reacted with a compound of the formula III

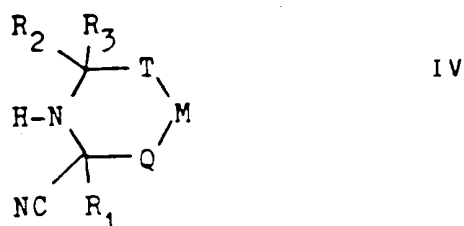


II

III

in which R denotes H or (C<sub>1</sub>-C<sub>4</sub>)alkyl, or

b) a compound of the formula II is reacted with an amine of the formula IV



or

c) where Y = 0, a compound of the formula I obtained under b) is hydrolyzed, and the compound obtained is converted, if appropriate, into its salt.

4. A herbicidal comprising a compound of the formula I of claim 1 or 2, or a salt thereof and an inert carrier.
5. The use of a compound of the formula I of claim 1 or 2, or a salt thereof, as a herbicide.
6. A process for controlling undesired plants, wherein an effective amount of a compound of the formula I, or a salt thereof, of claim 1 or 2 is applied to these plants or to the land used agriculturally.

DATED this 17th day of December 1987.

HOECHST AKTIENGESELLSCHAFT

EDWD. WATERS & SONS  
PATENT ATTORNEYS  
50 QUEEN STREET  
MELBOURNE, VIC. 3000.

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

**BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

☒ BLACK BORDERS

☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES

☐ FADED TEXT OR DRAWING

☐ BLURRED OR ILLEGIBLE TEXT OR DRAWING

☐ SKEWED/SLANTED IMAGES

☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS

☐ GRAY SCALE DOCUMENTS

☒ LINES OR MARKS ON ORIGINAL DOCUMENT

☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY

☐ OTHER: \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**

**THIS PAGE BLANK (USPTO)**